Nanoscale Roughness and Cliffs in Low-temperature Growth of a Copper Surface

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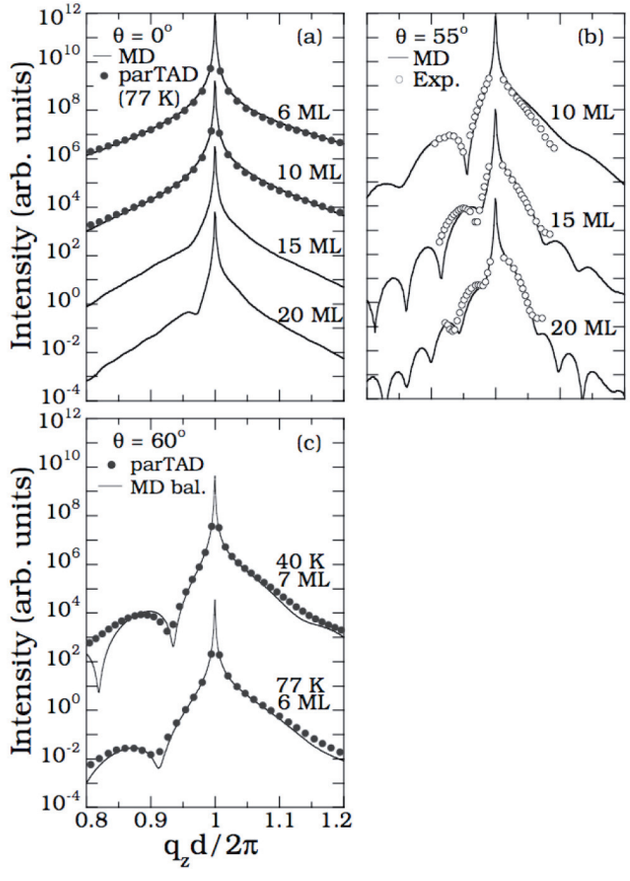

Fig. 1. X-Ray diffraction (XRD) patterns for copper films grown on Cu(100). a) synthesized XRD spectrum from molecular dynamics (MD) and ParTAD simulations at normal deposition angle. b) Comparison of experimental XRD pattern [4] with XRD pattern predicted from MD at deposition angle (from normal) of 55 deg. c) Comparison of XRD spectra predicted by MD and ParTAD at 60 deg. The ParTAD films, grown 100,000 times slower, show a softening of the XRD peaks due to the smoothing of the surface caused by the thermal diffusion events during deposition.

The process of vapor-deposited film growth, in which a crystal or film is grown one atom at a time as atoms arrive at the surface, can give rise to a surprisingly rich variety of surface morphologies [1]. Even for a simple metal like copper or silver, raising the temperature at which the growth is performed can give a surface that is smooth, then rough, then smooth, and then rough again. This effect results from the interplay of the deposition events with the various atomic diffusion and relaxation mechanisms that are active at a given temperature. The deposition angle and deposition rate also come into play, and having more than one atom type complicates matters further, so the overall complexity is daunting if one is trying to choose experimental conditions that will give a desired film morphology. It is thus valuable to have atomistic simulation tools with which we can probe and build an understanding at the microscopic level.

While the standard atomistic simulation approach, molecular dynamics (MD), is limited to time scales of less than a microsecond, the temperature accelerated dynamics (TAD) method developed here at LANL [2] offers a way to reach the time scales of milliseconds and seconds on which film growth takes place (a typical growth rate is of the order of a monolayer per second). TAD achieves this by raising the temperature of the system to cause more frequent attempted activated events, and then correcting the dynamics by allowing only those events that would have occurred at the desired lower temperature. We have used TAD to study film-growth problems in the past, but we have been limited to systems of roughly 1000 atoms or fewer due to the poor scaling of the TAD method with system size. This has precluded studying film growth problems where the feature size in the morphology is larger than a few times the lattice constant.

Recently, building on a development of Shim and Amar [3], we have developed an effective way to spatially parallelize a TAD simulation, so that we can reach a larger length scale at the same time as we extend the time scale. Although this ParTAD method [4] has additional overhead, it allows us to study systems many times larger than before, opening the way for studying more complex film growth systems, as well as many other processes in materials science that involve substantial time and length scales simultaneously.

A good example of a film-growth system that can benefit from atomistic interpretation is the low-temperature, off-axis growth of copper on Cu(100), recently studied experimentally by Miceli and coworkers [5-7]. Using X-ray diffraction analysis, they observed large compressive strains normal to the surface for films grown below T = 100K, from which they concluded that large concentrations of vacancies were incorporated into the films as they grew.

Using MD and ParTAD, combined with an accurate embedded atom method [8] interatomic potential for copper [9], we have simulated the growth of these films [10]. The X-ray diffraction curves synthesized from the atomistic simulations are in impressively good agreement with the experimental ones (see Fig. 1), but we find a striking difference in the interpretation of what gives rise to the compressive strain and the features in the spectrum. There are almost no bulk vacancies in the film, but we do find an anisotropic nanoscale roughness when the films are grown off-axis, as shown in Fig. 2. Moreover, we find a surprising tendency for the film to form perfectly vertical [100] walls, or "cliffs," as shown in Fig. 2d. The roughness arises from a combination of suppression of key activated events (due to the low temperature) that would otherwise smooth the film, and a shadowing effect due to the off-axis nature of the arriving atoms. Even at these low temperatures, thermally activated events do occur, but they do not strongly affect the film morphology—direct MD simulations of the growth on artificially fast time scales, allowing only a few picoseconds between successive depositions so that most activated events do not have time to occur, give films that are qualitatively similar (see the X-ray spectrum comparison in Fig. 1c). However, when the growth is simulated using kinetic Monte Carlo, a widely used model built on standard assumptions about the activated-event mechanisms, the result is a very different film morphology with nanoscale pyramids, so realistic atomistic detail is important in understanding this type of film growth process.

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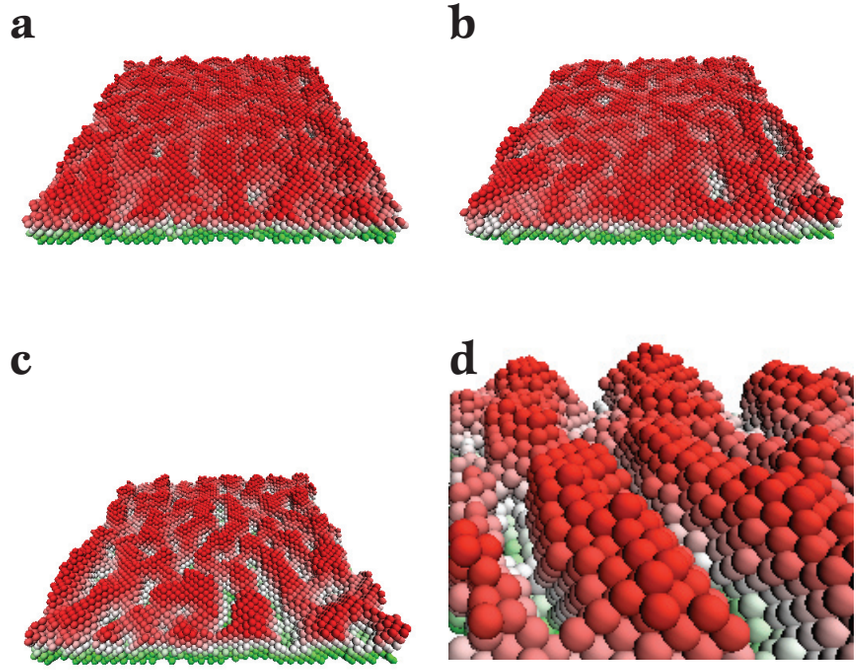

Fig. 2. Surface morphology for copper films grown to seven monolayer thickness on Cu(100) at T = 40K using ParTAD. The deposition rate is 5000 monolayers per second. a) Normal deposition incidence – note the smoothness. b) Deposition angle (from normal) 30 deg, and c) 60 deg. Note the increasing nanoscale roughness with increasing deposition angle (arrows show the direction of deposition) and the steep canyon walls at 60 deg. d) Close-up of c), showing completely vertical cliff walls.

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